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INCLUSION OF THE SORPTION HYSTERESIS PHENOMENON IN FUTURE DRYING MODELS. SOME BASIC CONSIDERATIONS♣

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ABSTRACT

The sorption hysteresis effect, i.e. different wood equilibrium moisture contents (EMCs) in desorption and adsorption for the same relative humidity, is well known. However, quantitative sorption isotherms, in the form of tables or analytical correlations, are almost always given as the average of the desorption and adsorption curves. Consequently most drying simulation models use these average curves, and does not take into account the sorption hysteresis phenomenon.

The equilibrium state of a wood sample is thus not a function of the relative humidity only, but depends on the moisture history also. This means that Fick's equations - with moisture content as a single driving force - are not valid any more. For a pure desorption process the state of the sample follows the desorption isotherm, but a problem arises when desorption is followed by adsorption - as for instance in the timber conditioning phase. It seems reasonable to assume that for each EMC point, on or between the desorption/adsorption isotherms, the moisture content change follows a unique path when the surrounding climate changes. This path - the so called scanning curve - does not need to be the same in desorption and adsorption. Some selected results and corresponding scanning curve suggestions are presented and discussed.

Drying models with the sorption hysteresis phenomenon included should be developed for the analysis of experimental data and more generally for use as an improved tool in practical applications.

Keywords: Modelling, sorption hysteresis, wood drying

INTRODUCTION

The sorption hysteresis is a well known phenomenon for wood. Hysteresis means in this context that the equilibrium moisture content (EMC) is different in desorption and adsorption processes. However, when quantitative sorption isotherms are presented in the literature (especially in handbooks such as Anon 1999 or Babiak 2007 normally only one curve is given, i.e. the average (AvEMC) of the desorption (DeEMC) and adsorption (AdEMC) curves. There are relatively few analytical expressions for wood (covering a wide temperature range) for the DeEMC and AdEMC curves separately (Hartley and Avramidis 2002, Krupinska *et al.* 2007). Perhaps due to this, many drying simulation models are based on the AvEMC curves for different temperatures. This introduces an error in the models, which to some extent has been handled by introducing correction factors.

In the following it will first be shown that neglecting the sorption hysteresis phenomenon in model work can have a substantial effect on model predictions. After that some of the main problems and new features connected with the introduction of hysteresis in modelling are pointed out. Finally some solutions to these problems are discussed.

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Is the sorption hysteresis important in modelling?

The interaction between a piece of wood and the surrounding climate (air temperature and humidity) is one of two main parts in all drying models. The other part is the moisture migration inside the wood. The external interaction includes transfer of heat (energy) as well as moisture (mass) to and from the wood surface. The heat and mass transfer are expressed with the following equations (Salin 2007).

$$\Phi / A = \alpha(T_\infty - T_s) \quad (1)$$

$$\dot{m}/A = \beta(c_s - c_\infty) \quad (2)$$

Φ/A = heat flux per unit area (W/m^2)
α = heat transfer coefficient (W/m^2/K)
$T\infty$, Ts = temperature of surrounding air and wood surface, resp. (K)
$\dot{m}/A$ = moisture flux per unit area (kg/m^2/s)
β = mass transfer coefficient (m/s)
c_∞ , c_s = vapour concentration of surrounding air and in equilibrium with the wood surface, resp. (kg/m^3)

Vapour concentration is used as the driving force in equation 2, but it can be discussed whether partial pressure would be a better choice. The term c_s gives the connection between surface moisture content (MC) and the vapour concentration in the air in equilibrium with the surface via the sorption curve. For a drying process the DeEMC curve should be used for determination of this connection.

As the transfer of heat and moisture occurs through the same air-side boundary layer, it seems reasonable that there should be a coupling between the transfer coefficients α and β. This is referred to as the analogy between heat and mass transfer, which with a good approximation can be expressed as:

$$\alpha / \beta = c_p \rho \quad (3)$$

where $c_p\rho$ is the volumetric heat capacity of the humid air in the boundary layer. Equation 3 is important as the mass transfer coefficient can be determined from the heat transfer coefficient which normally is more easily predicted. However, when drying model simulations have been compared with experimental data, it has frequently been found that equation 3 seems to overestimate the mass transfer coefficient (Salin 2007). As equation 3 represents a fundamental relationship, this deviation was not fully understood and correction factors were introduced into the models (Morén *et al.* 1992, Salin 1996, Hukka 1999, Hukka and Oksanen 1999).

However, the deviation can be explained by the fact that these models used the AvEMC sorption curves. This is illustrated by figure 1.

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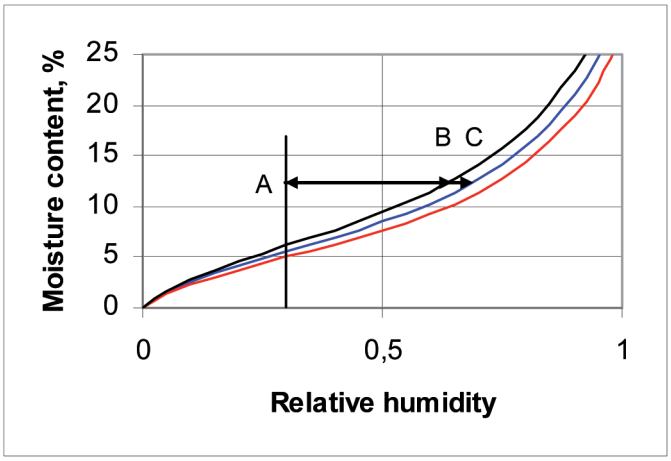

Figure 1. Example of a drying process in a sorption diagram where A represents the RH of the air and B the RH in equilibrium with the MC of the wood surface. In models the RH determined by the average sorption curve (C) is however often used. The curves are, from the top, desorption (DeEMC) average (AvEMC) and adsorption (AdEMC) curves.

In figure 1 the real driving force *(in an ideal uninterrupted drying process)* is represented by the distance A-B, which is shorter than the distance A-C used in the model. The correction factor needed is then AB/AC. As seen the correction factor becomes more important when the points B and C are close to the point A. This is in agreement with experimental findings regarding the correction factor (Morén *et al.* 1992, Salin 1996). In theory the factor can even be negative. It is further noticed that the correction factor is not a pure wood property, as it is dependent on the relative humidity (RH) of the surrounding air also. (RH is not a physically correct driving force, but as sorption diagrams traditionally use RH as a variable, it is used here for the purpose of illustration only) .

It seems that the use of AvEMC sorption curves instead of DeEMC curves in drying processes explain the observed deviations from the analogy between heat and mass transfer (Salin 2007). As it has turned out to be necessary to use correction factors in this context in present models, a much more reasonable solution would be to include the sorption hysteresis phenomenon directly in future models.

In drying of timber at sawmills, the distance A-B in figure 1 is normally relatively long, as rapid drying is preferred. For wooden material in buildings – both indoors and outdoors – the climate variation is normally much smaller and changes frequently from adsorption to desorption and vice versa. As noticed above, the correction factor becomes more important when the points A, B and C are close to each other. This means that the sorption hysteresis phenomenon becomes especially important in such building applications.

It can be argued that for ideal uninterrupted drying processes it is sufficient to use the DeEMC sorption curves instead of the AvEMC curves. However, the drying process includes an equalisation and/or conditioning phase, where the wood surface absorbs moisture. If so, the error is 'doubled' in this part. For large batch kilns it may also occur that timber on the leeward side absorbs moisture just after a fan reversal.

Based on these arguments, there is a need to include the sorption hysteresis phenomenon in drying models and as well in models for the interaction between climate and wood in buildings. There are complicated issues for implementing hysteresis into the models and some of the main points are summarized below.

Modelling sorption hysteresis

One important feature that is introduced when sorption hysteresis is included in the model is illustrated by the following scenario. Consider a completely dry piece of wood. One end of the wood is dipped into water for a short period of time and the MC in the end increases, i.e. to 20 %. Afterwards the wood is stored in a climate corresponding to an AvEMC of 10 %. The dry end MC will gradually approach 10 % but will stop at about 9 % due to the hysteresis effect (AdEMC ~9 %). In the same way the wet end of the piece will also approach 10 % but stops at about 11 % as DeEMC ~11 %. This situation is illustrated in figure 2. The interesting question is now; will there be a bound water migration from the higher MC towards the lower MC?

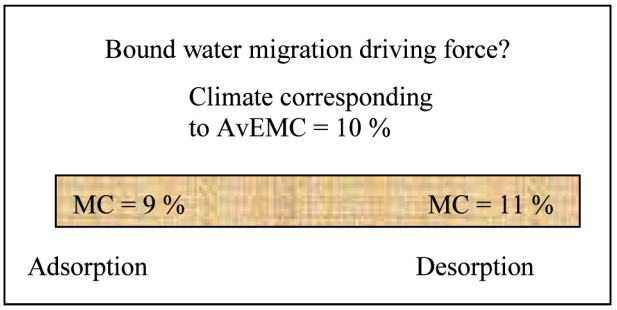

Figure 2. Scenario to illustrate the bound water driving force problem.

As both ends of the stick are in thermodynamic equilibrium with the same climate, they should be in mutual equilibrium, i.e. no moisture migration should occur. This shows that Fick's law with MC (or any other 'moisture concentration in wood' variable) is not valid any more when sorption hysteresis is taken into account. The state of the wood is not defined by the MC alone, but depends also on how this MC has been reached, i.e. the history of its moisture changes. This means that the driving force for bound water migration has to be changed. The logical solution is to use the state of the air in equilibrium with the wood as the driving force. This idea was suggested that vapour partial pressure is driving force (Bramhall 1979) and that chemical potential is driving force (Kawai *et al.* 1978, Stanish 1986). In addition to this change of driving force, it has to be established how the state point of the wood changes, i.e. in which direction will the state move, expressed as a point on or between the DeEMC and AdEMC curves. This is discussed later on.

In this context a second scenario is presented. Consider now a thin wood stick initially with an MC corresponding to the fibre saturation point (FSP). One end is dipped into water for a very long period of time so that this end eventually becomes completely saturated with water, i.e. an MC in the range 120-250 % depending on wood density. A flow of free water will occur due to capillary forces from the wet end towards the other end. The saturated end is in equilibrium with 100 % RH as there is free water at the surface. This proves that wood with an MC corresponding to FSP is not in equilibrium with 100 % RH, which however often is inaccurately stated. In addition many textbooks and handbooks present diagrams and tables where an EMC of about 30 % is given for 100 % RH. Kelvin's law that states that the vapour pressure above a curved liquid surface (meniscus) is lower than above a flat surface is important in this context. This imagined experiment brings of course the focus on the definition of the FSP. Equilibrium with 100 % RH is thus not a useful definition.

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Thus, of all the FSP definitions suggested (Stamm 1971) the most useful FSP definition is the point when the cell wall is water saturated but with no free water present. It is generally accepted that several mechanical wood properties – shrinkage/swelling, MOE, MOR, mechano-sorptive creep, etc. – depend on the amount of bound water but not on the amount of free water. There may be a transition zone and not a well defined transition point, but as these mechanical properties are very important for drying induced stress development as well as for shape deformations etc., this definition seems appropriate. Experimental determination of FSP values according to this definition constitutes a problem (as with many other FSP definitions). In addition, modelling moisture migration just below or above FSP is still a problem. From a drying modelling point of view it seems logical to extrapolate experimental sorption curves towards 100 % RH – using one of the many sorption models suggested in the literature (Vidal and Cloutier 2005, Babiak 2007) and define this point as "EMC for 100 % RH". In addition Kelvin's law should be neglected above FSP. However, it is not given that extrapolation of the DeEMC and AdEMC curves give the same FSP value, and this constitutes still a problem in the modelling procedure.

EMC is in most cases defined as the MC a piece of wood gradually attains when stored in a steady climate. It is then implied that the climate is not affected by the moisture flow to or from the sample. But there is another alternative to determine the EMC. If the sample is put into a small compartment, then the *climate* will gradually attain equilibrium with the sample and an EMC situation is the result in this case also. This method has actually been used for certain materials to indirectly determine the MC by measuring the RH in the compartment. In this case it is assumed that the moisture flow to or from the sample can be neglected in comparison with the total wood moisture, i.e. MC is not changed. Again, the result is not depending only on the sample MC, but also on how this MC has been attained, which is discussed in the next section.

An immediate question is whether these two alternatives represent the same basic EMC definition. It is perhaps not *a priori* self-evident, but by considering an intermediate situation where both air RH and sample MC change towards a mutual equilibrium, it is obviously correct. One should however be careful when comparing desorption from the wood and adsorption by the air (and vice versa). This is illustrated by the following example. Consider a cell wall that separates two air volumes (lumina) and that both the air and the wall are initially completely dry. Increase the RH on the 'left hand side' of the wall to a constant value. Then the wall absorbs moisture and the MC is increasing. Eventually the air RH on the 'right hand side' will start to increase as moisture is transferred through the wall. The air absorbs moisture from the cell wall, which looks like the wall would be in a desorption state and the air would follow the DeEMC curve, i.e. approach a lower RH on the 'right hand side'. But the cell wall MC is continuously increasing, i.e. the wall is actually in an absorption state and thus the AdEMC curve is followed and the RH will finally be the same on both sides. Both the wall and the 'right hand side' air are adsorbing moisture, although there is a moisture flow from the wall to the air. It should be kept in mind that the hysteresis phenomenon concerns the wood, not the air.

There are indications that the EMC and the hysteresis effect are dependent on the sample size (Shmulsky *et al.* 2001, Gjerdrum 2008). It is known from both experimental results and theoretical principles, that mechanical stress in the wood will influence the EMC. This has been suggested as an explanation, as very small samples should be free of stress. This is, in the author's opinion, not a very convincing explanation as tensile stress in the sample should be balanced by compression stress elsewhere – in the absence of external forces. Probably the sample size dependence is an artefact originating from another not fully understood phenomenon. Another astonishing result is the dependence of EMC on drying air velocity reported in (Shmulsky *et al.* 2002). Anyway, such phenomena have to be investigated further.

Scanning curves

For a hygroscopic material without sorption hysteresis, the state of the material will follow a single curve upon changes in the surrounding climate. For wood the situation is more complicated. In a pure drying process the state of the wood follows the desorption curve – but if the climate changes to an adsorption situation, then the state point will leave the desorption curve and move towards the adsorption curve. The question is now, along which transition curve will the point move? This transition curve is called a scanning curve (Cloutier and Fortin 1994, Peralta 1995, Frandsen 2007). One reasonable first approximation is to assume that the scanning curve is a nearly horizontal straight line between the boundary curves, as illustrated by the A-B-C-D sequence in the upper part of figure 3. This was among others proposed by (Time 1998, 2002) but she suggested also that a sloping curve, as in the lower part of figure 3, would reflect the real situation better.

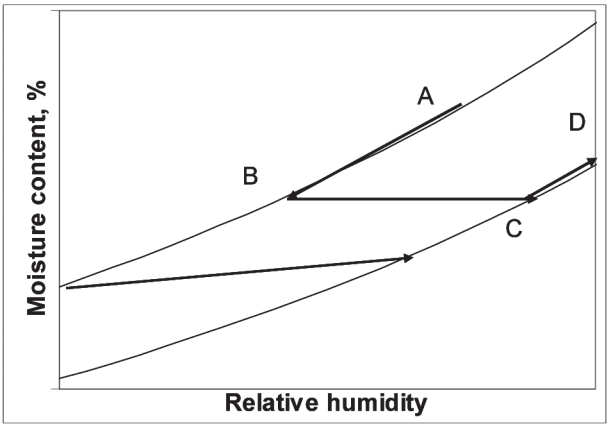

Figure 3. Examples of scanning curve alternatives.

In (Frandsen 2007, Frandsen and Damkilde 2000) it is assumed that for each point on or between the DeEMC and AdEMC boundary curves, there is a single scanning curve in the desorption direction and a single curve in the adsorption direction (not necessarily the same curve). This means that it doesn't matter how this point has been reached, i.e. it is assumed that history has no influence at this stage. Based on measurements found in the literature, (Frandsen 2007, Frandsen and Damkilde 2000) propose rather complicated expressions for the scanning curves. These expressions have no physical background, but are chosen to fit experimental data only. Examples of scanning curves are presented in figure 4 (adsorption) and figure 5 (desorption), calculated using the parameter values proposed.

It is seen in figures 4 and 5 that these scanning curves are far from horizontal. Further, a curve that starts from, for instance, the DeEMC boundary curve will approach the AbEMC boundary curve but never reach it. The same applies in the other direction.

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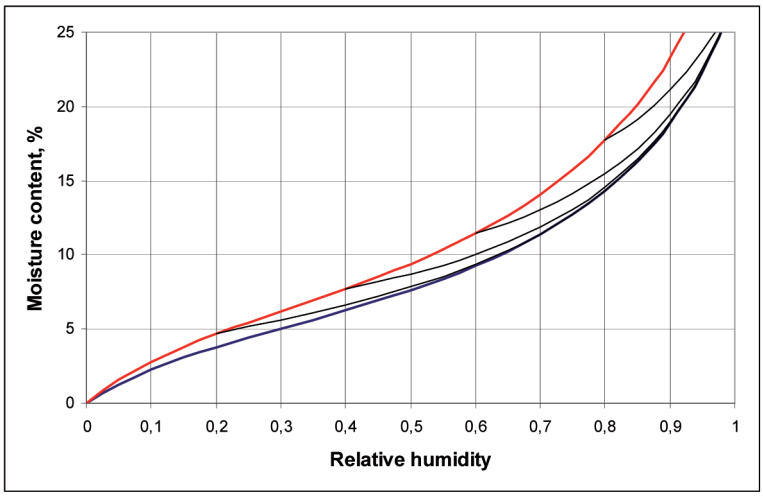

Figure 4. Example of adsorption scanning curves according to Frandsen (2007).

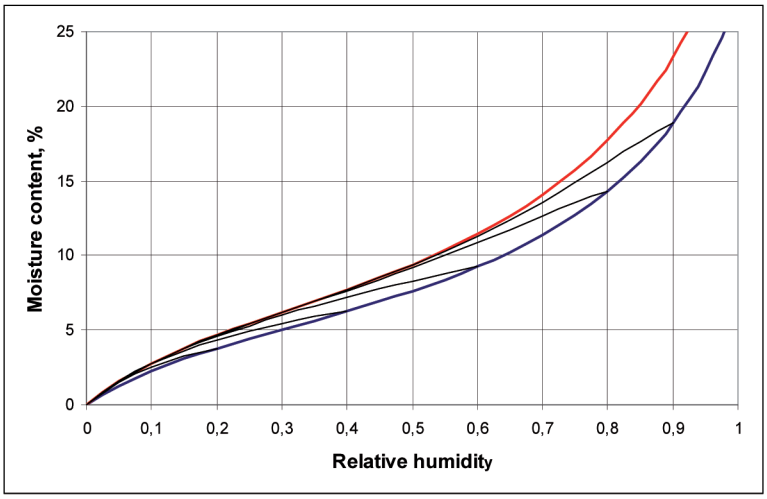

Figure 5. Examples of desorption scanning curves according to Frandsen (2007).

The scanning curve model proposed by (Frandsen 2007, Frandsen and Damkilde 2000) is relatively easily implemented in drying simulation software. Other scanning curve models have been suggested, for instance by using the 'independent-domain theory' (Peralta 1995), but this model is much more difficult to implement in simulations. As the amount of experimental data in this field is rather limited, it seems that the suggestions by (Frandsen 2007, Frandsen and Damkilde 2000) could be taken as a starting point for future model development. Such models should give relatively reliable predictions regarding the influence of sorption hysteresis in different situations.

A wood drying model with the sorption hysteresis phenomenon included has been presented in (Frandsen 2007, Frandsen and Svensson 2007) but it is not fully clear how the failure of MC as driving force has been accounted for. Very few other attempts in this direction have been presented (Ilic and Turner 1991, Cloutier and Fortin 1994) and in some cases the influence of approximations made, remains an open question.

CONCLUSIONS

It has been shown that there clearly is a need to include the sorption hysteresis phenomenon in future drying models, as well as in models for the interaction climate/wood in building environment. Some of the rather extensive problems connected to such an inclusion have been discussed and a few qualitative solutions have been presented.

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